Multimode Emissions from MEH-PPV Blended with Polystyrene Film Waveguides

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ABSTRACT— In the context of conjugated polymers, especially those of the poly (phenylenevinylene) (PPV) family, which are promising candidates as emission material in light emitting devices such as LEDs, field effect transistors and photovoltaic devices, we have, in the present study, prepared MEH-PPV [Poly [2-methoxy-5-(2'-ethyl-hexyloxy)-1,4-phenylen-evi-nylene] of low molecular weight and low poly-dispersity index. We discuss the thickness dependent fluorescence emission from the film waveguides fabricated with the above blends. Of the two peaks observed in this fluorescence spectrum, the shorter wavelength peak (562nm) is dominant in the range of lower thickness and as thickness is increased the longer wavelength peak (600nm) predominates. We also discuss the multimode laser emission from free standing MEH-PPV/Polystyrene film waveguides blends which show the nature of improvement in lasing behavior. The gain studies show that free standing film of MEH-PPV blended with Polystyrene can be used as a compact solid state laser.

KEYWORDS: Light emitting polymers, Multimode emission, Photoluminescence, film waveguide

I. INTRODUCTION

In recent years, conjugated polymers have emerged as an attractive new gain medium for lasers and optical amplifiers. Laser emission from conjugated polymers has been reported during the past few years [1]-[6].

As an advantage over organic laser dyes, the conjugated polymers can exhibit high photoluminescence quantum yields not only in solution but also in solid state. In the form of solid films, they can emit light apart from their capability of charge transport. These properties make them suitable for the realization of solid state electrically pumped lasers. Furthermore, excellent processability exists for the soluble conjugated polymers which lead to possibility of chemically tuning the emission wavelength. The absorption and fluorescence spectra are well separated, so that re-absorption of emitted light is minimum.

Among many of the conjugated polymers belonging to PPV family MEH-PPV ([Poly [2-methoxy-5-(2'-ethyl-hexyloxy) -1, 4-phen-ylen-evinylene]) is the most prominent one. The electronic and optical properties of MEH-PPV are highly sensitive to the conditions of chemical synthesis, photo-oxidation effects and film processing. In solid state films, there can be aggregation leads to excited interchain excitons which are responsible for majority of optical excitations [7]-[8]. The addition of polystyrene in MEH-PPV films induce a red shift in the absorption spectrum and reduce interchain interaction and increase the effective conjugation length of MEH-PPV main chain [9].

One of the parameters in the selection of conjugated polymers for laser media is poly dispersity index (PDI) which is a measure of

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the distribution of molecular mass in a given polymer sample. It indicates the distribution of individual molecular masses in a batch of polymers. Lower the PDI value, more advantageous for the conjugated polymer to function as efficient laser media. The PDI of MEH-PPV was reported to be greater than 1.5 [10]-[15]. We were able to bring down the PDI value as low as 1.38 because of the purification procedures we followed in each step.

In the present paper we report the observation of multimode laser emission from a transversely pumped free-standing film of MEH-PPV/PS blend. Since the film is free standing, air is the surrounding media on both sides of the film, the reflections from the lateral faces of the sample provide the optical feedback for laser action. This was evident from the observed dependence of lasing mode-spacing on the film thickness. The leaky mode emission leads modeling of the film waveguide as a planar micro cavity-like behavior due to Fabry–Perot effects. We also discuss the thickness dependence of fluorescence emission from the film waveguides.

II. EXPERIMENTS

Details of the synthesis and chemical characterization of MEH-PPV are explained elsewhere [16]. MEH-PPV was diluted in 10ml of 1, 2 dichlorobenzene and blended with PS. The free standing films of different thicknesses were prepared by Tape casting method. When the solvent was fully evaporated, high-quality free-standing films could be peeled off from the glass substrate. The lateral faces of the films obtained were of good optical finish such that no further polishing was required. The films were cut into the size 4 cm × 2 cm. The concentration of MEH-PPV from was varied from 2mg to 10mg and repeated the same experiment. The film quality was found to be highly dependent on the solvent used. We tried different solvents such as Tetrahydrofurane, Dichloromethane, toluene, and chloroform. But the films were not sufficiently transparent and the surface was non uniform. Blending of MEH-PPV with PMMA instead of with PS does not provide good quality. Then also the film quality was very poor.

The absorption and fluorescence spectra were recorded using UV-VIS spectrophotometer (Jasco V-570) and Cary Eclipse Spectrofluorimeter respectively. The laser emission was observed using the set up reported previously for Amplified Spontaneous Emission (ASE) [17]. Along with a cylindrical lens, pump beam in the form of a stripe was used for limiting the level of illumination. The samples were transversely pumped using 7 ns pulses from a frequency doubled Nd:YAG laser (532 nm, 10 Hz). For varying the pump energy from 2mJ/pulse to 9mJ/pulse a set of calibrated neutral density filters were used. The beam was focused into a narrow stripe length and studies were carried out by varying the stripe length of 2mm, 4mm and 8 mm. The emission from the sample was collected by an optical fiber and directed to a 0.5 m spectrograph (SpectraPro-500i) coupled with a cooled CCD array. The distance between the collecting fiber and the waveguide edge was fixed to an optimum value of 1 cm.

III. RESULTS AND DISCUSSION

Absorption and fluorescence spectra of MEH-PPV/Polystyrene blended film are shown in Fig. 1.

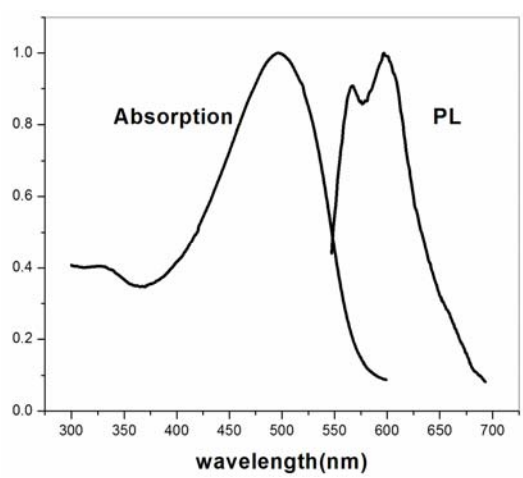

Fig. 1 Absorption and PL spectra of MEH-PPV/PS blend free standing film

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The overlap between the spectra is not much so that self absorption of fluorescence emission is minimum. The peak of the absorption band was observed at 497nm and the shape remained the same irrespective of the concentration and thickness is clear from the in Fig. 2.

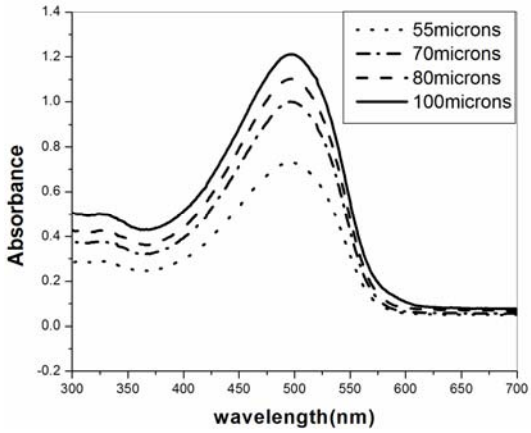

Fig. 2 Variations of absorption spectra of free standing film of MEH-PPV/PS blend with respect to thicknesses from 55microns to 100microns as given in the text.

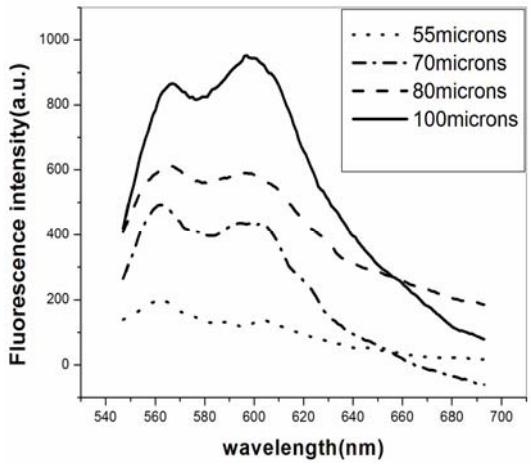

Fig. 3 Variations of fluorescence emission spectra with respect to film thickness.

From Fig. 3 it can be seen that, the nature of fluorescence emission spectra depend on the film thickness. The emission spectra show twin peaks at 562nm and 600nm. The two peaks are due to the two vibrational transitions 0-0 and 0-1 bands [18]. At smaller thicknesses the shorter wavelength peak (562nm) the intensity of which is got reduced gradually with higher wavelength peak (600nm) becoming dominant. We carried out the above experiment at various concentration of MEH-PPV and the observations detailed above did not change.

Results clearly show a redistribution of intensity corresponding to two vibrational modes as film thickness is changed. The relative intensity of the 0-0 band is very sensitive to film thickness. The relative intensities of 0-0 and 0-1 bands change with respect to film thickness. The dual peaks are present up to an optimum concentration above which the shorter wavelength emission is suppressed. (See table 1). Similar results in respect of dual amplified spontaneous emission were observed in MEH-PPV dissolved in organic solvents [16].

Table 1 Ratio of peak intensities of the twin peaks obtained in fluorescence spectra as a function of film thickness

Thickness (μm)	P_{00}/P_{01}
55	1.46
70	1.13
80	1.04
100	0.91

There exists an energy transfer between shorter wavelength and longer wavelength bands depending upon various parameters like temperature, concentration and thickness [19].The solid solution of MEH-PPV contains conjugation segments in the separated state while there are many conjugation segments in aggregated states in MEH-PPV films. The aggregation of the polymer chain supports Forster type energy transfer for the excited states. This Forster type energy transfer of the excited states in conjugated polymers occurs very fast with a fine scale of pico-seconds [20]-[22]. Owing to the rapid energy transfer, the excitons can easily migrate to low energy sites including quenchers, allowing a single quencher site to put out the emission from hundreds of chromophores in the conjugated polymer [23]-[24]. The guest chromophore is diluted by the host polystyrene which suppresses the interchain interactions leads to enhance the PL efficiency [9]. The addition of PS into MEH–PPV improves the thermal stability of polymer thin film which is

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advantageous in device point of view since it reduces the sensitivity of device performance with processing conditions.

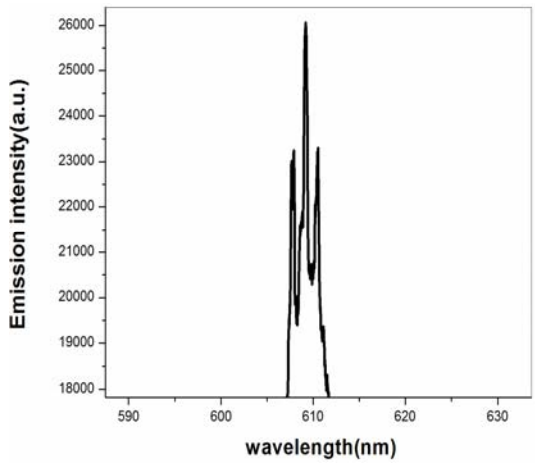

Fig. 4 Multimode emission from transversely pumped MEH-PPV/PS blend free standing film of thickness 80micrometer pumped at 2mJ/pulse.

When the film was transversely pumped by the pulsed laser of 2mJ of energy the laser modes were observed at a mode separation of 1.3nm as shown in Fig. 4. Normally an external feedback is required for Laser emission. Here, there were no external mirrors to provide the feedback. When a free-standing film is pumped, the lateral faces will act as mirrors. This will give rise to Fabry–Perot type optical cavity where the film thickness acts as the length of the cavity. In the case of a thin stripe excitation, the stimulated emission occurs in the direction along the stripe [25]. The propagation through the guiding gain medium and the stimulated emission occurring in the medium are repeated by the feedback at the lateral faces. This results in high gain although the reflections at the lateral faces are much smaller than that of conventional cavity mirrors.

The fine structure pattern in the spectra shown in Fig. 4 can be attributed to the axial modes of the Fabry–Perot cavity formed by two surfaces of the planar waveguide. In other words the planar waveguide can be thought of as being formed by a number of serially connected Fabry–Perot etalons. This can be verified from the mode spacing as shown below.

For the multimode lasing spectrum, the wavelength spacing between different modes is given by:

$$\Delta\lambda = \lambda^2 / 2nL \qquad (1)$$

where λ is the average lasing wavelength. Substituting for the average lasing wavelength λ as 609 nm, the mode spacing $\Delta\lambda$ obtained in our studies as 1.3 nm with L *a*s the length of the resonator cavity which is the thickness of the film, measured as 80micron, we can evaluate the refractive index as 1.7 which is the refractive index of MEH-PPV. Typically, polymeric films have refractive indices in the range of 1.6–1.8. The ratio of concentration of MEH-PPV and polystyrene in the above film was 0.005:1 and the energy of laser pulse was 2mJ. When the energy is increased to 3.3mJ, single mode is evolved while the others are suppressed as shown in Fig. 5. This is due to mode competition which leads to the survival of mode with maximum gain alone.

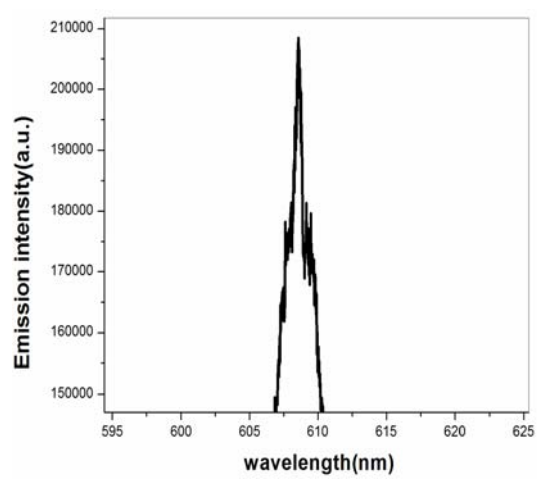

Fig. 5 Multimode emission when pump energy was increased to 3.3mJ/pulse, central mode is dominant.

At laser power of 5.5mJ and above we observed structures similar to Random lasing. Even in perfect polymer films, there might be substantial scattering due to impurities and fluctuations in the film's density and thickness. This may produce weak

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backscattering resonances that may be the reason for the structure shown in Fig. 6.

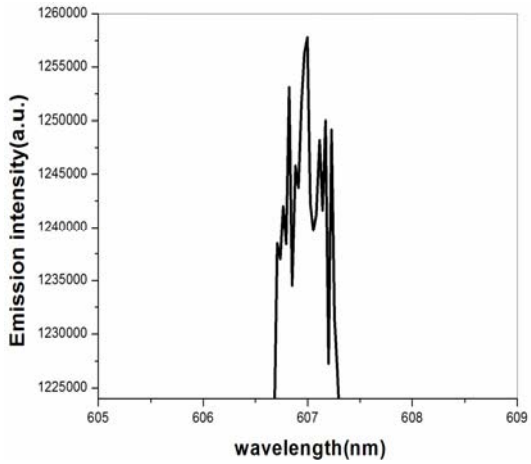

Fig. 6 Multimode emission is changed to Random lasing at 5.5mJ/pulse

In a waveguiding structure light is channeled through a series of total internal reflections between the boundaries of different refractive index media. In this way light propagation can be limited to small dimensions corresponding to the light wavelength. This light confinement together with high electric field strength associated with the pump beam largely enhances the probability of stimulated emission processes by the increase of the effective interaction length. For expanding the light guiding effect over a larger distance, the waves reflected from successive boundaries must interfere constructively. Due to this restriction, various patterns of constructive interference are guided along the channel and these give rise to a discrete number of waveguide modes. The total number of modes depends not only on the refractive indices, but also on the thickness of the waveguide region. Below a cut-off thickness modes are no longer supported and light propagation is solely due to leaky modes that suffer continuous propagation loss.

As the thickness of the film was increased, a red shift of the peak was noted because of the variation of concentration of MEH-PPV with film thickness. As the concentration is increased the aggregate formation is more, which is the lower band gap species compared to the isolated polymer chain [8]. Aggregation happens more in aromatic solvents such as Dichlorobenzene which interacts mainly with the aromatic backbone of the polymer chain. There are two vibronic transitions in PL spectrum in which the shorter wavelength peak was completely suppressed, when pumped with pulsed laser in free standing films.

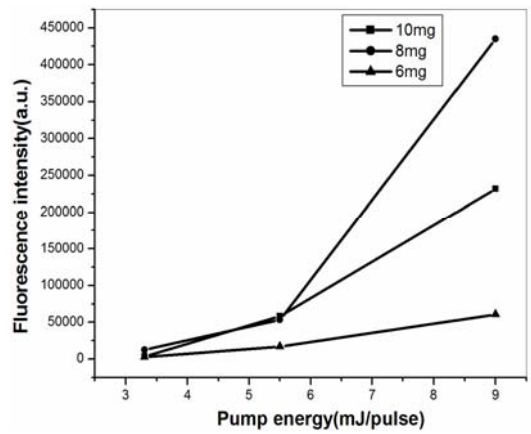

Fig. 7 Variation of fluorescence intensity with pump energy of films with different concentrations

At 8mg concentration, we observe minimum FWHM of 4.1nm at 5.5mJ pump energy. Amplified spontaneous emission (ASE) is the dominant mechanism in films since the output pulse energy increases exponentially with stripe length until it reaches saturation while the broad emission spectrum narrows [26]. As the pump energy is further increased, FWHM is increased due to gain saturation.

We also studied the dependence of peak fluorescence intensity on concentration and film thickness. The fluorescence intensity is found to enhance abruptly above threshold pump power as shown in Fig. 7. The presence of threshold is an indication of optical amplification. Maximum amplification occurs at 8mg concentration. The thickness dependence of FWHM at 4mg concentration is shown in Fig. 8.

As the thickness of the film increases, the size of the gain medium increases which enhances the lasing and reduces the FWHM. For the film of 80 micron thickness we observe

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minimum FWHM which increases at higher thicknesses because of gain saturation.

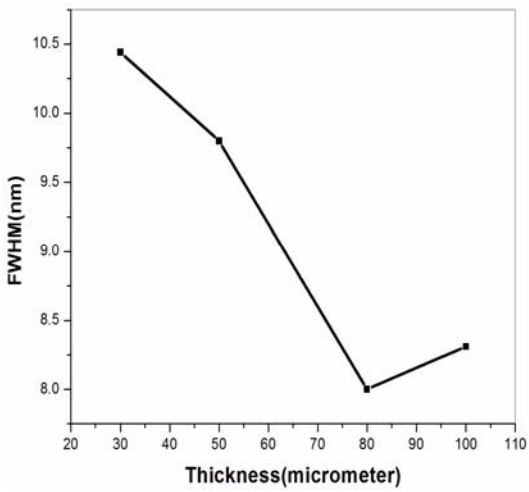

Fig. 8 The thickness dependence of FWHM at 4mg concentration

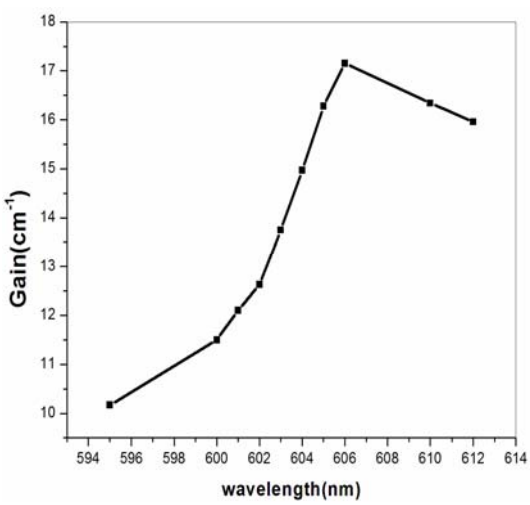

Fig. 9 Gain spectrum of freestanding films of MEH-PPV/PS blends

Gain was calculated using the standard definition of laser gain as given by Shank [27].

$$g = \frac{2}{l}\ln\left[\frac{I_l}{I_{l/2}} - 1\right] \quad (2)$$

where l is the length of the pumped region, I_l is the fluorescence intensity propagating along the Z-axis through the stripe length of l, and $I_{l/2}$ is the intensity propagating through half of the stripe length, $l/2$ .The gain of ASE was found to be maximum, 17.2cm^{-1} at 606nm, for a concentration of 10mg, thickness 75 micrometer and energy of 5.5mJ/pulse. This value is comparable with those of efficient laser dyes like Rhodamine6G. The gain spectrum in Fig. 9 shows that the MEH-PPV/PS blended film can be used as a solid state laser media.

IV. CONCLUSION

We have prepared MEH-PPV [Poly[2-methoxy-5-(2'-ethyl-hexyloxy)-1,4- phenyl-enevinylene] using a modified procedure and purified several times in each steps of synthesis. Film waveguides based on MEH: PPV/PS blends were fabricated by tape casting technique. The photoluminescence spectra of the film waveguide show twin peak emission nature of which varies with the thickness of the film. The shorter wavelength peak is dominant at lower thicknesses and as the thickness was increased that peak was decreased and longer wavelength peak was dominant. This is due to the effect of greater concentration quenching of shorter wavelength in comparison with that of the higher wavelength emission. The overlap of absorption and PL spectrum also affect the shorter wavelength at higher concentration. We observed the multimode laser emission from transversely pumped free standing film waveguides of MEH-PPV/PS blends. Reflections from the lateral faces provided the optical feedback and together with the guidance through the gain medium give rise to intense narrow emission lines. The presence of polystyrene matrix reduces the interchain interactions within the medium. The gain studies show that free standing film of MEH-PPV blended with Polystyrene can be used as a compact solid state laser media.

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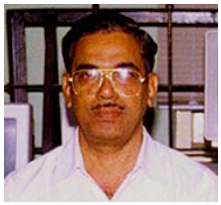

Prof. C.P. Girijavallabhan received PhD degree from Kerala University in 1971. He did his postdoctoral research at Southampton University in the United Kingdom. He was also a visiting professor at Franhaufer Institute, Freiberg, Germany. Currently he is Emeritus Professor at ISP and CELOS. He is the founder director of International School of Photonics. His research interests include lasers, laser matter interactions, optoelectronics, molecular physics and solid state physics. He is the member of the governing council of the Nuclear Science Centre (NSC), President of Photonics Society of India and a fellow of the Optical Society of India. He was the Vice President of the Indian Laser Association. He has published more than 200 research papers in international journals.

Prof. P. Radhakrishnan has received Ph.D. degree from Cochin University of Science and Technology in 1986. He has been a lecturer at

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the Cochin College from 1979 to 1988. Presently he is a Professor and Director of International School of Photonics. He is also the Director of the Center of Excellence in Lasers and Optoelectronic Sciences, Cochin University of Science and Technology. His research interests include laser technology, laser spectroscopy, and fiber optic sensors. He is the present President of the Photonics Society of India and life member of Indian Laser Association, Indian Association of Physics Teachers, and the Indian Physics Association.

Dr. V.P.N. Nampoori is the Professor of Photonics in International School of Photonics, Cochin University of Science and Technology. He took his PhD in Physics in the field of Molecular Spectroscopy. At the present, he is working in topics related to Lasers, Photonics and Nonlinear Dynamics. Dr. Nampoori is one of the members of the core team to establish a well developed Laser and Photonics Laboratory in CUSAT. Now this is one of the leading laboratories in Lasers and Photonics in India. He also designed the course structure for Integrated M.Sc. Photonics Course as a part of the Center of Excellence in Lasers and Optoelectronic Sciences. Dr. Nampoori has published more than 300 research papers and supervised about 15 PhDs in lasers, laser spectroscopy and fiber optics. Significant contributions of Dr Nampoori are in the area of Fiber Optic Sensors, Nonlinear Optics and Photonic materials. Recently his group has developed a fiber optic amplifier which works in the visible region of the light spectrum. Dr. Nampoori is also interested in the field of Science Education through e-learning platform, popularization of science by writing popular articles in Malayalam and English periodicals.

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